IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

T. IMAI et al

SERIAL NO. 09/840,878 GROUP ART UNIT: 1754

FILED: April 25, 2001 EXAMINER: E. M. JOHNSON

FOR: IRON COMPOUND CATALYST FOR INHIBITING GENERATION OF DIOXIN AND INCINERATION PROCESS OF MUNICIPAL SOLID WASTE USING THE SAME

DECLARATION UNDER 37 C.F.R. 1.132

HONORABLE COMMISSIONER OF PATENTS & TRADEMARKS

WASHINGTON, D.C. 20231

SIR:

Now comes Toshiki MATSUI, a citizen of Japan, and a resident of 1-4-605, Miyake 2-chome, Saeki-ku Hiroshima-shi, Hiroshima-ken, Japan, who declares and says that:

1. I graduated from the Department of Ferment Engineering, Faculty of Engineering, Hiroshima University in March 1985.

2. I am currently employed by TODA KOGYO CORPORATION since April, 1985.

3. I am familiar with the work related to U.S. Patent Application, Serial No. 09/840,878, and am a co-inventor of the U.S. Patents: No. 5,993,536 and No. 6,276,287.

4. Under my control and supervision the following experiments were conducted:

Experiment 1

3.5 liters of a 1.0 mol/liter aqueous ferrous sulfate solution were charged into a reactor (5 liters). After heating the content of the reactor to 45°C passing therethrough at an air flow rate of 15 liters/min, 1.0 liter of a 1.4 mol/liter aqueous sodium hydroxide solution (the amount of alkali was 0.2 equivalents based on the ferrous iron (Fe^{2+})) was added to the reactor, and then heated to 50°C to conduct a reaction and the reaction was continued until deceasing the pH value to not more than 3. Next, after heating the obtained suspension to 80°C, a 1.4 mol/liter aqueous sodium hydroxide solution were dropped to the suspension while maintaining the pH of 4 to react the whole Fe ions, thereby producing goethite particles. Thereafter, the obtained goethite particles were successively subjected to filtration, washing with water, drying and pulverization.

The thus obtained goethite particles had an average particle size of 0.48 μm, a BET specific surface area of 15 m^2/g, a phosphorus content of 0.002 % by weight, a sulfur content of 0.06 % by weight and a sodium content of 0.07 % by weight, and the conversion percentage of carbon monoxide into carbon dioxide at a temperature of 250°C according to the following specified evaluation method was 19 %.

Experiment 2

The goethite particles obtained in the above Experiment 1 were heat-treated at 780°C for 10 hours in air, thereby obtaining hematite particles.

The thus obtained hematite particles had an average

particle size of 0.27 μm, a BET specific surface area of 5.0 m^2/g, a phosphorus content of 0.002 % by weight, a sulfur content of 0.07 % by weight and a sodium content of 0.08 % by weight, and the conversion percentage of carbon monoxide into carbon dioxide at a temperature of 250°C according to the following specified evaluation method was 18 %.

Experiment 3

2.0 liters of a 1.0 mol/liter aqueous ferric chloride solution and 3.0 liters of a 3.7 mol/liter aqueous sodium hydroxide solution (the amount of alkali was 1.8 equivalents based on the ferric iron (Fe^{3+})) were charged into an autoclave (5 liters). The resultant mixture was maintained at 40°C for 6 hours and then was reacted at 180°C for 4 hours, thereby producing goethite particles. Thereafter, the obtained goethite particles were successively subjected to filtration, washing with water, drying and pulverization.

The thus obtained goethite particles had an average particle size of 1.5 μm, a BET specific surface area of 1.0 m^2/g, a phosphorus content of 0.002 % by weight, a sulfur content of 0.01 % by weight and a sodium content of 0.08 % by weight, and the conversion percentage of carbon monoxide into carbon dioxide at a temperature of 250°C according to the following specified evaluation method was 15 %.

The properties in the examples were measured by the following methods.

(1) The average particle size of the iron oxide particles or iron oxide hydroxide particles was expressed by the value

measured from an electron micrograph.

(2) The specific surface area of the iron oxide particles or iron oxide hydroxide particles was expressed by the value measured by a BET method.

(3) The contents of phosphorus and sodium contained in the iron oxide particles or iron oxide hydroxide particles were expressed by the values measured by an inductively coupled plasma atomic emission spectrometer (SPS-4000 Model, manufactured by Seiko Denshi Kogyo Co., Ltd.).

(4) The content of sulfur contained in the iron oxide particles or iron oxide hydroxide particles were expressed by the value measured by a Carbon-Sulfur Analyzer (EMIA-2200 Model, manufactured by Horiba Seisakusho Co., Ltd.).

(5) As to catalyst property of the iron oxide particles or iron oxide hydroxide particles catalyst for inhibiting the generation of dioxin, the catalytic activity was expressed by a conversion percentage of carbon monoxide into carbon dioxide by measuring the concentration of carbon dioxide produced when 2.8×10^{-4} mol of iron oxide particles (α-Fe_2O_3) obtained by heat-treating the iron compound catalyst in air at a temperature of 800°C for 15 minutes, were instantaneously contacted with 6.1×10^{-7} mol of carbon monoxide at a temperature of 250°C at an SV of 42,400

h^{-1} in an inert gas atmosphere using a pulse catalytic reactor.

Here, the "SV" means a space velocity, and is expressed by the value obtained by dividing a flow rate of the reaction gas by a volume of the catalyst. The SV is represented by an inverse number of time (h^{-1}).

The pulse catalytic reactor used comprises a reactor portion and a gas chromatography portion which is constituted by Gas Chromatography GC-16A (manufactured by Shimazu Seisakusho Co., Ltd.).

The evaluation method used herein was conducted by referring to methods described in the literatures (e.g., R. J. Kobes, et al, "J. Am. Chem. Soc.", 77, 5860(1955) or "Experimental Chemistry II-Reaction and Velocity" edited by Japan Chemistry Institute and published by Maruzen, Tokyo (1993)).

The results are shown in the Table below.

Table

	Properties of iron compound catalyst for inhibiting generation of dioxin		
	Average particle size of iron, cobalt and iron oxide (μm)	BET specific surface area (m^2/g)	Phosphorus content (wt.%)
Our invention	0.02-1.0	0.2-100	≤0.02
Experiment 1	0.48	15	0.002
Experiment 2	0.27	5	0.002
Experiment 3	1.5	1	0.002

Table (continued)

	Properties of iron compound catalyst for inhibiting generation of dioxin		
	Sulfur content (wt.%)	Sodium content (wt.%)	Catalyst property (conversion percentage of carbon monoxide into carbon dioxide at 250°C) (%)
Our invention	≤0.1	≤0.2	≥18
Experiment 1	0.06	0.07	19
Experiment 2	0.07	0.08	18
Experiment 3	0.01	0.08	15

Remarks

As seen from the above, the conversion of carbon monoxide into carbon dioxide of are Experiments 1 and 2 are 19 % and 18 %, respectively. On the contrary, the conversion of carbon monoxide into carbon dioxide of are Experiment 3 is 15 % which is inferior to those of Experiments 1 and 2 which are within a scope of our invention.

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6. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

7. Further, deponent saith not.

Date: November 18, 2003

Toshiki Matsui

Toshiki MATSUI